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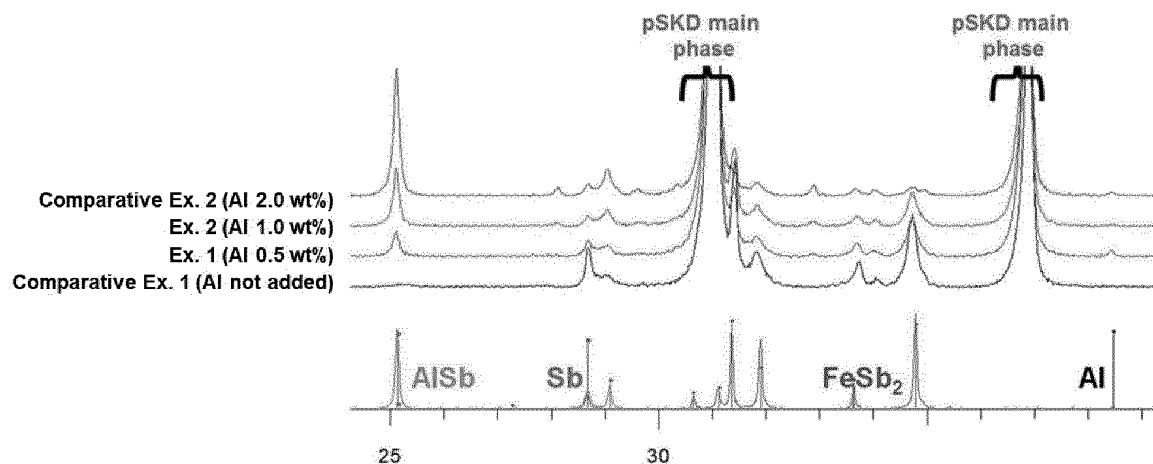
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(54) **THERMOELECTRIC MATERIAL AND THERMOELECTRIC ELEMENT COMPRISING SAME**

(57) The thermoelectric material according to the present invention minimizes the content of components that degrade thermoelectric performance and thus can be usefully used in thermoelectric devices including the same.

[FIG. 1]



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Description**[Technical Field]**5 **CROSS-REFERENCE TO RELATED APPLICATION(S)**

[0001] This application claims the benefit of the filing date of Korean Patent Application No. 10-2018-0066825 filed with the Korean Intellectual Property Office on June 11, 2018, the entire content of which is incorporated herein by reference.

10 **[0002]** The present invention relates to a thermoelectric material minimizing the content of components that degrade thermoelectric performance, and a thermoelectric device including the same.

[Background Art]

15 **[0003]** Recently, with interest in the development and saving of alternative energy being heightened, investigation and research on efficient energy conversion materials are actively conducted. In particular, research on thermoelectric materials, which are thermoelectric energy conversion materials, is being accelerated.

[0004] These thermoelectric materials are metal or ceramic materials having a function to directly convert heat to electricity or inversely convert electricity to heat, and have the advantage that power can be generated even in the absence of moving parts when a temperature difference is applied. After the discovery of their thermoelectric phenomena, namely Seebeck effect, Peltier effect, and Thomson effect at the beginning of the 19th century, these thermoelectric materials have been developed so as to have a high thermoelectric performance index as semiconductors have progressed since the late 1930s.

25 **[0005]** Meanwhile, as next-generation materials having thermoelectric energy conversion characteristics, skutterudite compounds are researched. Skutterudite compounds have abundant potential as materials that are expected to improve their thermoelectric energy conversion characteristics by the decrease of lattice thermal conductivity.

[0006] Meanwhile, a method for producing a thermoelectric material containing skutterudite generally includes three stages, wherein a raw material is first heated under various conditions in accordance with the chemical composition of the target thermoelectric material, and subjected to heat treatment to synthesize an ingot-shaped material. This is pulverized and classified to produce a powder, which is then sintered at high temperature and high pressure to produce a thermoelectric material.

[0007] By the way, the production method including the synthesis, pulverization, classification and sintering as described above is expensive to apply in a continuous process, and therefore, in the industrial production, in consideration of economic efficiency, the thermoelectric material powder is first mass-produced as described above, and subsequently, the powder is sintered to produce the thermoelectric material. In other words, the production process of the powder is more economical as mass production becomes possible, and the sintering process of the powder has a long process time due to the high temperature process characteristics. Thus, it takes a considerable amount of time between the powder mass production process and the sintering process and therefore inevitably stores the powder for a long time.

40 **[0008]** Meanwhile, the present inventors have found that the performance of the thermoelectric material finally produced greatly depends on the storage conditions of the skutterudite powder produced by the above-mentioned method. In particular, as described in detail below, it has been confirmed that materials that degrade the performance of the thermoelectric material are found in the thermoelectric material produced while the skutterudite powder is stored for a long time.

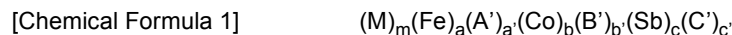
45 **[0009]** In this regard, the present inventors have conducted an intensive research on a method for preventing the performance of the thermoelectric material from degrading even when producing the thermoelectric material after the powder is stored for a long time as described above, and as a result, have found that when using the thermoelectric material and the production method thereof as described below, the above-mentioned problems are solved, thereby completing the present invention.

50 **[DETAILED DESCRIPTION OF THE INVENTION]****[Technical Problem]**

55 **[0010]** It is an object of the present invention to provide a thermoelectric material minimizing the content of components that degrade thermoelectric performance, and a thermoelectric element including the thermoelectric material.

[Technical Solution]

[0011] In order to achieve that above objects, one embodiment of the present invention provides a thermoelectric material comprising a p-type skutterudite material of the following Chemical Formula 1, greater than 0 wt% and 7.5 wt% or less of AlSb, and greater than 0 wt% and 20 wt% or less of FeSb₂:



in Chemical Formula 1,

M is at least one of element selected from the group consisting of S, In, Nd, Pr, Ce, Yb, La, Sr, Ba, Ca, Sm, Eu, and Gd, $0 < m < 1.5$,

A' is at least one of element selected from the group consisting of Ni, Mn, Tc, and Pd, $1 < (a + a') \leq 4$, and $0 \leq a' < 1$,

B' is at least one of element selected from the group consisting of Ni, Ru, Os, Ir, and Pt, $0 < (b + b') < 3$, and $0 \leq b' < 1$,

C' is at least one of element selected from the group consisting of Sn and Te, and $11 < (c + c') < 13.5$, and $0 \leq c' < 1$.

[0012] As used herein, the term 'p-type skutterudite material' refers to a compound represented by Chemical Formula 1 as defined above, and corresponds to a main component of a thermoelectric material in the present invention.

[0013] The p-type skutterudite-based thermoelectric materials as described above is generally produced by a method for producing the thermoelectric materials including the steps of heating raw materials under various conditions in accordance with the chemical composition, subjecting them to heat treatment to produce an ingot-shaped material, pulverizing and classifying the resultant material to produce a powder, and then sintering the powder at high temperature and high pressure. In industrial production, the skutterudite material is produced by mass-producing the powder and then sintering it for economic reasons. In this process, sintering is proceeded after the powder is stored for a long time. The long-term storage causes a problem that the performance of the thermoelectric material is greatly deteriorated.

[0014] In particular, the "p-type skutterudite" of the present invention contains Fe and Sb elements. Although not theoretically limited, the performance degradation of the thermoelectric material as described above is attributable to the generation of FeSb₂ due to long-term storage, as in the Examples and Comparative Examples of the present invention. In addition, the "p-type skutterudite" of the present invention also contains Co element, and thus the FeSb₂ may also exist in the form of (Fe/Co)Sb₂. Therefore, FeSb₂ described herein means also including (Fe/Co)Sb₂.

[0015] Therefore, according to the present invention, in the production of the thermoelectric material, a small amount of Al is added and sintered together during the sintering of p-type skutterudite powder, and thereby a small amount of AlSb is generated in the thermoelectric material. As a result, it is possible to reduce the generation of FeSb₂ which degrades the performance of a thermoelectric material.

[0016] Preferably, m is 0.5 to 1.0. Also, preferably, the M is Nd.

[0017] The a is preferably $2 \leq a \leq 4$, and most preferably 3.

[0018] The b is preferably $0.5 \leq b \leq 2$, and most preferably 1.

[0019] The c is preferably $11.5 \leq c \leq 12.5$, and most preferably 12.

[0020] The a', b', and c' mean the amounts of elements that are substituted and entered into Fe, Co, and Sb sites, respectively. The a', b', and c' are each less than 1, and when a', b', and c' are 0, it means that there is no element substituted.

[0021] Meanwhile, the content of the AlSb and FeSb₂ mean being based on the total mass of the thermoelectric material described above. For example, when AlSb and FeSb₂ are respectively included at 5 wt%, it means that the thermoelectric material contains 5 wt% of AlSb, 5 wt% of FeSb₂, and 90 wt% of the p-type skutterudite material.

[0022] As described below, the AlSb is generated by adding Al during the production of the thermoelectric material, and is contained in an amount of greater than 0 wt% and 7.5 wt% or less in the thermoelectric material of the present invention. Although not theoretically limited, as Al is sintered together, Al and Sb are combined to form AlSb, and thereby, the generation of FeSb₂ is suppressed and as a result, it is possible to prevent the performance of the thermoelectric material from deteriorating.

[0023] As the generation of AlSb increases, the generation of FeSb₂ lowers, and as the generation amount of AlSb increases, the performance of the thermoelectric material is improved. However, when the generation amount of AlSb is excessively large, the performance of the thermoelectric material is deteriorated. Therefore, preferably, the generation amount of AlSb is preferably 7.0 wt% or less, 6.5 wt% or less, 5.0 wt% or less, 4.5 wt% or less, or 4.0 wt% or less in the thermoelectric material. Further, preferably, the generation amount of AlSb is 0.5 wt% or more, 1.0 wt% or more, or 1.5 wt% or more in the thermoelectric material.

[0024] As AlSb is generated, the generation amount of FeSb₂ decreases. Preferably, it is 20 wt% or less, 19 wt% or less, 18 wt% or less, 17 wt% or less, 16 wt% or less, 15 wt% or less, 14 wt% or less, 13 wt% or less, or 12 wt% or less in the thermoelectric material.

[0025] Another embodiment of the present invention provides a method for producing the thermoelectric material described above, comprising the steps of:

- 1) mixing M, Fe, A', Co, B', Sb, and C" at a molar ratio of m: a: a': b: b': c: c", and then heating and heat-treating the mixture to produce an ingot;
- 2) pulverizing the ingot produced above to produce a powder; and
- 3) mixing the pulverized powder with an Al powder at greater than 0 to 1.5 wt% relative to the weight of the powder and then sintering it:

wherein M, A', B', C', m, a, a', b, b', c and c' are as defined above.

[0026] Step 1 is a step of mixing the components of the thermoelectric material and heating and heat-treating the mixture to produce an ingot.

[0027] Preferably, the heating temperature is 600 to 1400 °C. Preferably, the heating time is 10 to 200 hours. Preferably, the heat treatment temperature is 500 to 800 °C. Preferably, the heat treatment time is 10 to 200 hours. Thereby, the components of the thermoelectric material are finally melted to prepare an ingot-shaped material.

[0028] Step 2 is a step of pulverizing the ingot produced in step 1 to produce a powder.

[0029] The powder may be pulverized so as to have a particle size of 100 μm or less, and a classification step may be added as necessary. As the pulverization/classification method and apparatus therefor, the methods and apparatuses used in the field of inorganic materials can be applied without limitation.

[0030] Step 3 is a step of sintering the pulverized powder produced in step 2.

[0031] As described above, step 2 and step 3 are difficult to apply in a continuous process, in industrial production, and therefore, a thermoelectric material is produced by mass-producing a powder as in steps 1 and 2, and then sintering the powder as in step 3. This greatly affects the performance of the thermoelectric material depending on the conditions under which the powder is stored between step 2 and step 3.

[0032] However, in the present invention, by adding Al to the powder and sintering together as in step 3, it is possible to minimize the content of components that degrade performance in the thermoelectric material.

[0033] The addition amount of Al is a factor for determining the content of AlSb and FeSb₂ in the above-mentioned thermoelectric material. Therefore, by adjusting the addition amount of Al, it is possible to adjust the content of AlSb and FeSb₂. Preferably, the addition amount of Al is 0.1 wt% or more, 0.2 wt% or more, 0.3 wt% or more, 0.4 wt% or more, or 0.5 wt% or more, and 1.4 wt% or less, 1.3 wt% or less, 1.2 wt% or less, 1.1 wt% or less, or 1.0 wt% or less, relative to the powder produced in step 1.

[0034] The sintering may be carried out at a temperature of about 500 to 900°C using a spark plasma sintering. Preferably the sintering temperature is 600 to 680°C. In addition, the sintering time is preferably 1 minute to 600 minutes at a pressure of 0.1 to 100 MPa.

[0035] Still another embodiment of the present invention provides a thermoelectric device comprising the above-mentioned thermoelectric material.

[ADVANTAGEOUS EFFECTS]

[0036] As described above, the thermoelectric material according to the present invention minimizes the content of components that degrade thermoelectric performance and thus can be usefully used for thermoelectric devices including the same.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0037] FIG. 1 shows an XRD graph of thermoelectric materials produced in Examples and Comparative Examples according to the present invention.

[DETAILED DESCRIPTION OF THE EMBODIMENTS]

[0038] Hereinafter, preferred examples are provided to facilitate understanding of the present invention. However, the following examples are merely provided for a better understanding of the present invention, and the scope of the present invention is not limited thereto.

Comparative Example 1

(Step 1)

- 5 **[0039]** High purity raw materials, Nd, Fe, Co and Sb were weighed in a glove box at a molar ratio of 0.9: 3.0: 1.0: 12.1, put into a graphite crucible and then charged into a quartz tube. In the case of Sb, it was added by a molar ratio of 0.1 for volatilization. The inside of the quartz tube was sealed in a vacuum state. Then, the raw material was heated at 1000 to 1200°C and kept constant temperature state in the furnace for 24 hours. Next, the quartz tube was naturally cooled to room temperature to form an ingot, which was then again kept constant temperature state in a furnace at 600 to 750°C for 120 hours and subjected to heat treatment. The heat-treated ingot material was pulverized and classified into a powder having a particle size of 75 μm or less.

(Step 2)

- 15 **[0040]** The powder produced in step 1 was stored in a glove box with an oxygen concentration of 1 ppm or less for 6 months.

(Step 3)

- 20 **[0041]** 7 g of the powder produced in step 2 was put into a 0.5-inch SUS mold and cold-pressed at a pressure of 0.5 to 1 ton to prepare a pSKD pellet. The prepared pellet was put into a 0.5-inch carbon mold and subjected to a high-temperature pressurization at 650°C and 50 MPa for 10 minutes through a spark plasma sintering (SPS) device to prepare a 0.5-inch pSKD sintered body.

Example 1

- 25 **[0042]** Powders were prepared in the same manner as in steps 1 and 2 of Comparative Example 1. 0.5 wt% of Al was mixed with 7 g of the powder. This was put into 0.5-inch sus-mold and cold-pressed at 0.5 to 1 ton to prepare a pSKD pellet. The prepared pellet was put into a 0.5-inch carbon mold and subjected to a high-temperature pressurization at 650°C and 50 MPa for 10 minutes through a Spark plasma sintering (SPS) device to produce a 0.5-inch pSKD sintered body.

Example 2

- 35 **[0043]** The pSKD sintered body was produced in the same method as in Example 1, except that 1.0 wt% of Al was used instead of 0.5 wt% of Al.

Comparative Example 2

- 40 **[0044]** The pSKD sintered body was produced in the same method as in Example 1, except that 2.0 wt% of Al is used instead of 2.0 wt% of Al in step 3 of Example 1.

Experimental Example 1: XRD Analysis

- 45 **[0045]** The phases of the P-type skutterudite thermoelectric materials prepared in Examples and Comparative Examples were analyzed by an X-ray diffractometer (XRD) and the results are shown in FIG. 1.

[0046] As shown in FIG. 1, AlSb was not observed in the case of Comparative Example 1 in which Al was not added. Further, it was confirmed that the degree of generation of FeSb_2 was lowered depending on the degree of addition of Al.

- 50 **[0047]** Meanwhile, after XRD measurement, the composition and content of the chemical materials included were calculated through Rietveld refinement (RWP <10), and the results are shown in Table 1 below.

Experimental Example 2: Performance evaluation of thermoelectric materials

- 55 **[0048]** The sintered bodies produced in the Examples and Comparative Examples were processed into right-angled columns having a size of 3 mm $\times$ 3 mm $\times$ 12 mmH, and then electrical conductivity (EC) and Seebeck coefficient (S) were measured using ZEM3 (Ulbac) and LSR3 (Linseis) instruments. For comparison of the representative values of electrical characteristics, the comparative evaluation was performed by calculating the average values of the measurement evaluations at 100, 200, 300, 400, and 500°C.

[0049] In addition, the sintered bodies produced in the Examples and Comparative Examples were processed into a round column with a size of 2 mmT x 0.5 inch Φ , and the thermal diffusivity was evaluated using a LFA457 (Netzsch) instrument. The thermal conductivity (TC) was calculated from the measured thermal diffusivity, the apparent density of each sintered body, and the specific heat calculated by Dulong-Petit law. For comparison of the representative values of the heat transfer characteristics, a comparative evaluation was performed by calculating the average value of the thermal conductivity measurement evaluations at 100, 200, 300, 400, and 500°C.

[0050] The measurement results are shown in Table 1 below.

[Table 1]

	Addition amount of Al	Composition	EC (Scm^{-1})	S (μVK^{-1})	PF ($\mu\text{Wcm}^{-1}\text{K}^{-2}$)	TC ($\text{Wm}^{-1}\text{K}^{-1}$)	ZT
Comparative Ex. 1	(Not added)	$\text{Nd}_{0.9}\text{Fe}_{3.0}\text{Co}_{1.0}\text{Sb}_{12}$ FeSb_2 19.8 wt% AlSb 0 wt%	1162	112.3	14.58	2.76	0.306
Ex. 1	0.5 wt%	$\text{Nd}_{0.9}\text{Fe}_{3.0}\text{Co}_{1.0}\text{Sb}_{12}$ FeSb_2 12 wt% AlSb 1.8 wt%	1090	125.7	17.13	2.51	0.394
Ex. 2	1.0 wt%	$\text{Nd}_{0.9}\text{Fe}_{3.0}\text{Co}_{1.0}\text{Sb}_{12}$ FeSb_2 10 wt% AlSb 4.0 wt%	1063	128.1	17.35	2.46	0.410
Comparative Ex. 2	2.0 wt%	$\text{Nd}_{0.9}\text{Fe}_{3.0}\text{Co}_{1.0}\text{Sb}_{12}$ FeSb_2 3.7 wt% AlSb 9.7 wt%	1224	118.5	17.03	2.81	0.355

[0051] As shown in Table 1, as the addition amount of Al increased, the generation amount of FeSb_2 showed a decreasing tendency and the generation amount of AlSb showed an increasing tendency.

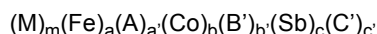
[0052] In addition, as shown in Examples 1 and 2, when the addition amount of Al was 0.5 wt% and 1.0 wt%, the thermoelectric material performance showed an increasing tendency. However, when the addition amount of Al was 2.0 wt% as in Comparative Example 2, the thermoelectric material performance was reduced, such as a decrease in the ZT coefficient. This is not limited in theory, but it is believed that the generation of AlSb increases which thus adversely affects the thermoelectric material.

Claims

1. A thermoelectric material comprising:

a p-type skutterudite material of the following Chemical Formula 1,
greater than 0 wt% and 7.5 wt% or less of AlSb, and
greater than 0 wt% and 20 wt% or less of FeSb_2 :

[Chemical Formula 1]



in Chemical Formula 1,

M is at least one of element selected from the group consisting of S, In, Nd, Pr, Ce, Yb, La, Sr, Ba, Ca, Sm, Eu, and Gd,

$0 < m < 1.5$,

A' is at least one of element selected from the group consisting of Ni, Mn, Tc, and Pd,

$1 < (a + a') \leq 4$, and $0 \leq a' < 1$,

B' is at least one of element selected from the group consisting of Ni, Ru, Os, Ir, and Pt,

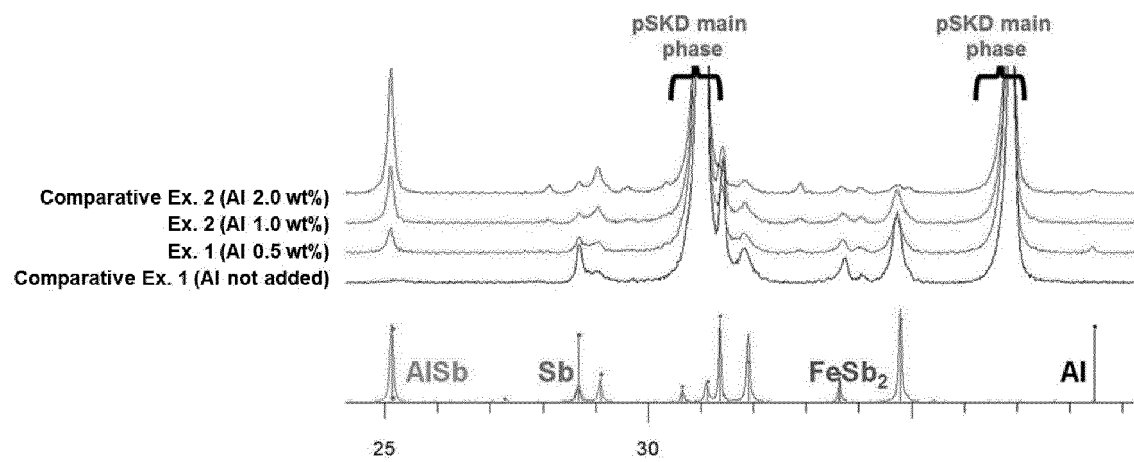
$0 < (b+b') < 3$, and $0 \leq b' < 1$,

C' is at least one of element selected from the group consisting of Sn and Te, and

$11 < (c+c') < 13.5$, and $0 \leq c' < 1$.

- 5 **2.** The thermoelectric material according to claim 1, wherein m is 0.5 to 1.0.
- 3.** The thermoelectric material according to claim 1, wherein the AlSb contains in an amount of 1 to 5 wt%.
- 4.** The thermoelectric material according to claim 1, wherein the FeSb₂ contains in an amount of 5 to 15 wt%.
- 10 **5.** A method for producing the thermoelectric material of any one of claims 1 to 4, the method comprising the steps of:
 - 1) mixing M, Fe, A', Co, B', Sb, and C' at a molar ratio of m:a'a':b':c':c', and then heating and heat-treating the mixture to produce an ingot;
 - 15 2) pulverizing the ingot produced above to produce a powder; and
 - 3) mixing the pulverized powder with an Al powder at greater than 0 to 1.5 wt% relative to the weight of the powder and then sintering it;wherein M, A', B', C', m, a, a', b, b', c and c' are as defined in any one of claims 1 to 4.
- 20 **6.** The method of claim 5, wherein the heating temperature of step 1 is 600 to 1400°C.
- 7.** The method of claim 5, wherein the heating treatment temperature of step 1 is 500 to 800°C.
- 25 **8.** The method of claim 5, wherein the addition amount of Al in the step 3 is 0.5 to 1.0 wt% relative to the weight of the powder.
- 9.** The method of claim 5, wherein the sintering temperature is 500 to 900°C.
- 30 **10.** A thermoelectric device comprising the thermoelectric material of any one of claims 1 to 4.

[FIG. 1]



INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2019/005996

A. CLASSIFICATION OF SUBJECT MATTER

H01L 35/18(2006.01)i, H01L 35/22(2006.01)i, H01L 35/16(2006.01)i, H01L 35/34(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

H01L 35/18; H01B 1/02; H01L 35/02; H01L 35/14; H01L 35/34; H01L 35/22; H01L 35/16

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models: IPC as above

Japanese utility models and applications for utility models: IPC as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS (KIPO internal) & Keywords: thermoelectric, skutterudite, powder, thermal treatment, sintering

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	KR 10-2017-0082982 A (LG CHEM, LTD.) 17 July 2017 See claim 1; and figure 1.	1-10
A	KR 10-2018-0057079 A (KOREA NATIONAL UNIVERSITY OF TRANSPORTATION INDUSTRY-ACADEMIC COOPERATION FOUNDATION) 30 May 2018 See paragraphs [0013]-[0020]; and figure 1.	1-10
A	KR 10-1469759 B1 (AGENCY FOR DEFENSE DEVELOPMENT) 08 December 2014 See paragraphs [0032]-[0051]; and figure 1.	1-10
A	US 2016-0141481 A1 (BOARD OF TRUSTEES OF MICHIGAN STATE UNIVERSITY) 19 May 2016 See paragraphs [0039]-[0057]; and figure 1.	1-10
A	US 2010-0319746 A1 (SNYDER, G. Jeffrey et al.) 23 December 2010 See paragraphs [0058]-[0081]; and figures 1-7.	1-10

☐ Further documents are listed in the continuation of Box C.
 ☒ See patent family annex.

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Date of the actual completion of the international search

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Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.

PCT/KR2019/005996

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